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Acoustic laser cleaning of silicon surfaces

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ABSTRACT We investigate the detachment of small particles from silicon surfaces by means of acoustic waves generated by laser-induced plasma formation at the back side of the sample. It is demonstrated that sufficiently high acoustic intensities can be reached to detach particles in the submicron regime. In order to study this "acoustic laser cleaning" in more detail, we have developed an interference technique which allows one to determine the elongation and acceleration of the surface with high temporal resolution, the basis for an analysis of the nanomechanical detachment process, which takes place on a temporal scale of nanoseconds. We find that the velocity of the detaching particles is significantly higher than the maximum velocity of the substrate surface. This indicates that not only inertial forces, but also elastic deformations of the particles, resulting from the acoustic pulse, play an important role for the cleaning process.

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1 Introduction

Adhesion phenomena are omnipresent in everyday life, but in spite of their importance, the underlying mechanisms are not yet fully understood, due to the complexity of the surfaces in contact, their structure and their interactions. This not only holds for the adhesion of biological objects like cells [1] or even animals like spiders and geckoes [2], where progress in the microscopic understanding has been achieved only recently, but is also true for, at first sight, much simpler examples like dust particles on flat surfaces. The adhesion of unwanted small particles poses serious problems in micro- and nanotechnology and is one of the main sources of malfunction, e.g., of semiconductor chips. For this reason understanding the mechanisms of adhesion is also crucial for the development of new cleaning schemes for particle removal from surfaces.

One of these schemes, being developed complementary to conventional ultrasonic techniques, is laser cleaning [3]. The basic concept in its simplest form, the so-called dry laser cleaning (DLC), is to heat the sample by a short laser pulse, which gives rise to a rapid expansion of the substrate. According to this concept inertial forces acting on the particles during the deceleration phase of the substrate surface can become sufficiently large to overcome the adhesion forces, and the particles will detach [4]. Vice versa, one might consider this technique not only for cleaning, but also as a means for a determination of adhesion forces, provided the time-dependent position of the substrate surface is known with sufficient accuracy. This dynamic approach appears as an alternative to the quasi-static measurement of adhesion forces acting on small particles by means of atomic force microscopy [5].

A direct application of this dynamic concept for determining adhesion forces is hampered, however, by the fact that in the standard DLC process further mechanisms come into play. In addition to the thermal expansion, local ablation of the substrate due to enhancement of the incident laser intensity by the optical near field of the particles occurs [6]. In this work we present a method in which these complications are avoided by irradiating not the front, but the back side of the sample. An acoustic pulse is generated, which propagates through the sample, leading to a momentary elongation of the surface without the simultaneous presence of an optical field in the vicinity of the particle. We describe below how the displacement can be measured with an accuracy of 0.1 nm and a temporal resolution of 0.2 ns and demonstrate that the inertial forces reached in this way are sufficient for an acoustic removal of test particles from silicon wafer surfaces by means of "acoustic laser cleaning".

2 Experimental setup

The pulsed laser source used in our experiments for the generation of the acoustic pulses was a frequency-doubled Nd:YAG laser with a pulse length of 10 ns at 532 nm and a Gaussian profile. The laser was focused to a 0.54 mm FWHM spot at the back side of the samples, 3.05 mm thick boron doped Si slabs with (111) orientation. The front side of the samples was either carefully cleaned for the measurements of the surface displacement (Figs. 2–4) or covered with test particles for the investigation of the acoustic cleaning efficiency (Figs. 5 and 6). As test particles we used colloidal polystyrene spheres with a diameter of 840 nm, which were distributed uniformly across the substrate by means of a spin coating pro-

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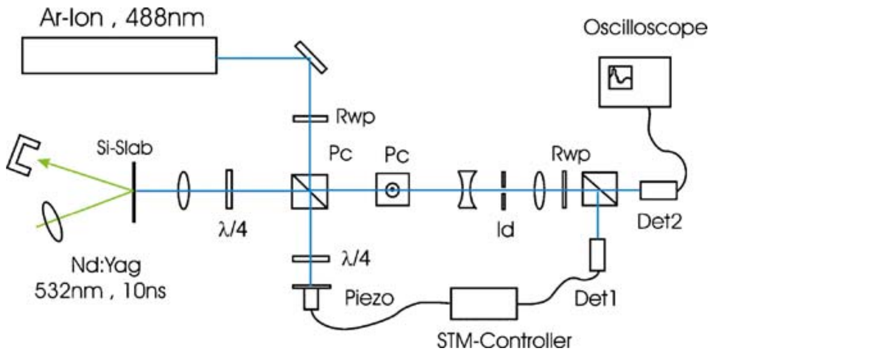

FIGURE 1 Experimental setup of the modified Michelson interferometer. Pc: polarizing cube, Id: iris diaphragm, Rwp: retarding waveplate

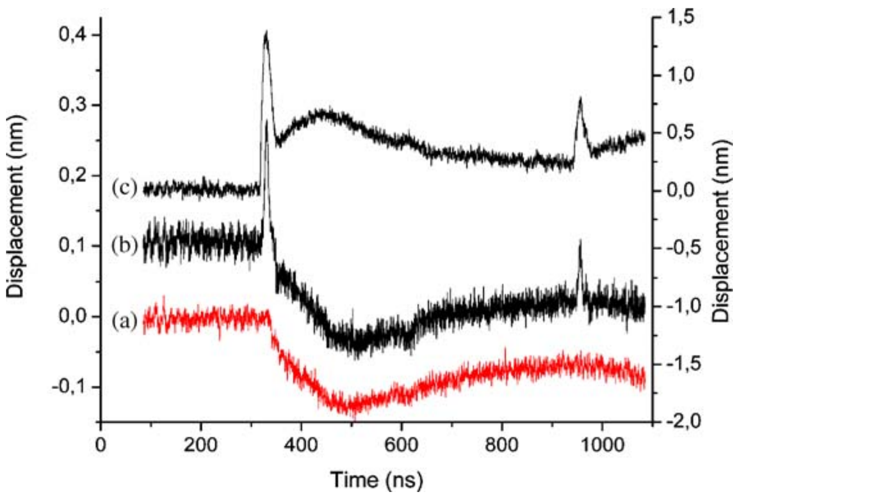

FIGURE 2 Surface displacement of a 3.05 mm thick (111) Si slab after exciting the back side of the sample with a laser pulse at time $t = 0$. Trace (a) (*left scale*): incident fluence 768 mJ/cm^2; trace (b) (*left scale*): 1.17 J/cm^2; trace c (*right scale*): 3.2 J/cm^2. For clarity, trace (b) has been shifted by 0.1 nm

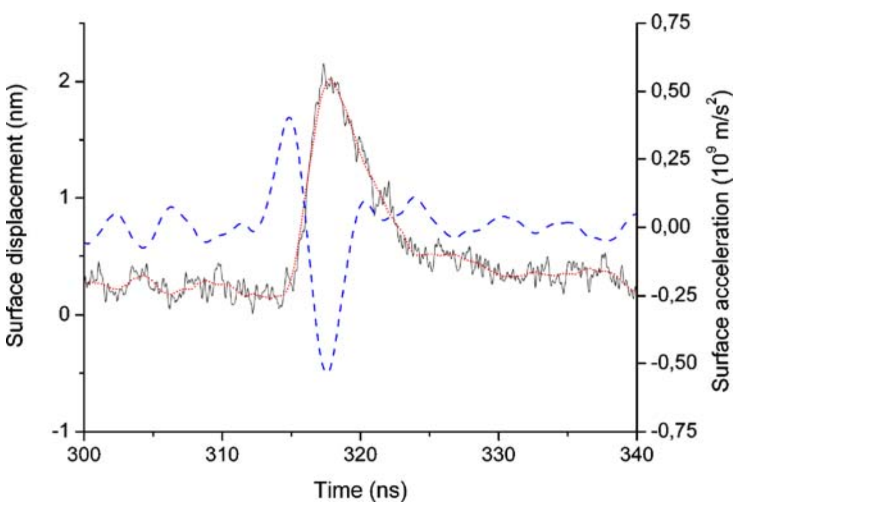

FIGURE 3 Longitudinal acoustic wave through a 3.05 mm thick (111) silicon wafer at 1.7 J/cm^2. The surface displacement is first fitted (*dotted line*) and from the fit the corresponding surface acceleration (*dashed line*, *right scale*) is derived

cess [7]. Whether or not these particles were removed from the sample surface by the acoustic pulse was determined by light scattering. For this purpose we used a He-Ne probe laser (632.8 nm) to illuminate the particles residing on the surface and a photomultiplier to register the scattered light. Any detachment of particles results in a decrease in scattering intensity. In addition, the samples were inspected afterwards ex situ with a dark field microscope, where particle detachment showed up as a dark spot (Fig. 5).

In order to detect the time-resolved displacement of the reflecting sample surface, we used a modified Michelson interferometer, as described in [8, 9] (Fig. 1). In our set-up the interferometer is stabilized by a piezoelectrically driven mirror in the reference arm. The adjustment is obtained by a common scanning tunneling microscope control unit. This leads to a stabilization of the interferometer up to 1 kHz, suitable for eliminating temperature fluctuations and slow vibrations, whereas faster mismatches between the two arms will not be corrected. Thus the optical output of the interferometer, registered with a fast photodiode (Det2 in Fig. 1), allows one to measure surface displacements with frequencies higher than 1 kHz. Full width at half maximum of our detector (Hamamatsu GA4176) is 200 ps as tested with a fs light pulse. The light source of the interferometer is an Ar-ion laser at a wavelength $\lambda_{int} = 488$ nm, single longitudinal mode and an output of 1.5 W cw. In contrast to a common interferometer set-up, we use a polarizing beam splitter cube in order to minimize reflections on the beam splitter surfaces, to control different reflectivities of the mirror materials and to avoid the reflection of the inverse interference pattern into the laser. To get the reflected light from both arms into the detection arm the quarter-wave plate rotates the polarization of the waves by 90° after two passes. Both reflected and orthogonally polarized beams of the two arms are brought to interference by a second beam splitter cube, as the cube transmits only those parts of the beams with the same polarization. From the measured intensity I in the center of the interference pattern the displacement Δd of the surface is obtained, using

$$I = \frac{I_{max}}{4R_0}\left[R_{Ref} + R_{Si} + \sqrt{R_{Ref}R_{Si}} \times \cos\left(\frac{4\pi\,\Delta d}{\lambda}\right)\right], \qquad (1)$$

where I_{max} is the maximum intensity, and R_{Ref} and R_{Si} are the reflectivities of the reference mirror and the silicon sample. The displacement resolution achieved with the interferometer is 0.1 nm for single shot experiments and

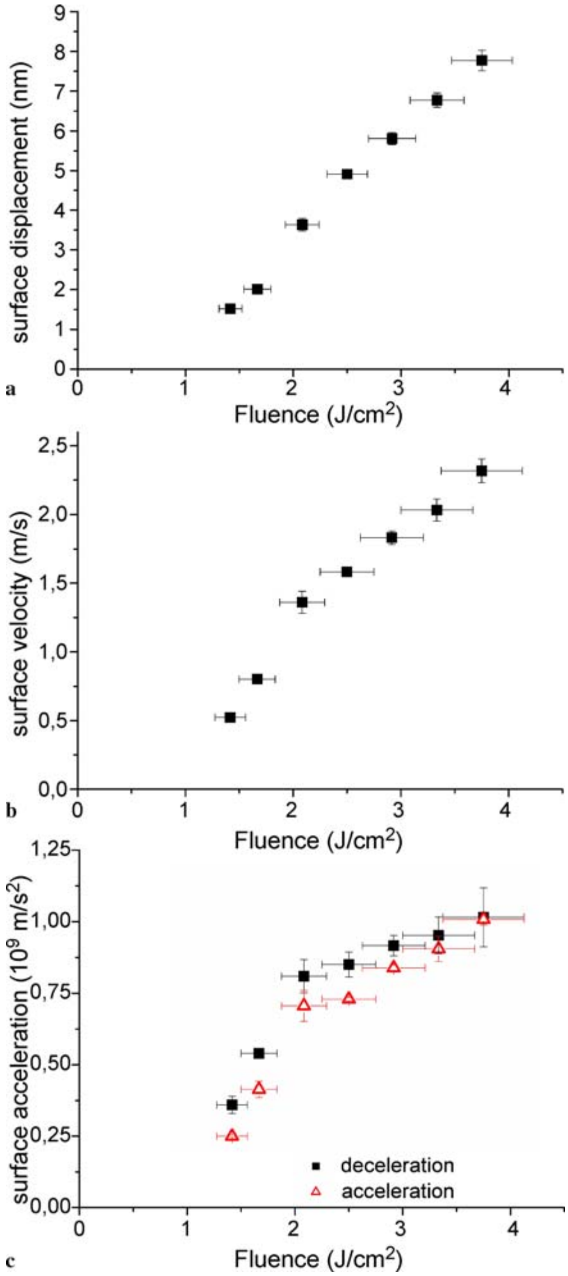

FIGURE 4 Response of the silicon surface due to the laser excited acoustic wave. (**a**) Maximum surface displacement after the ignition of a plasma on the opposing side. Furthermore the maximum surface velocity (**b**), acceleration and deceleration (**c**) of the silicon surface resulting from the ignited plasma are shown

0.02 nm in multi-shot measurements, with a lateral resolution of 50 μm.

3 Results and discussion

3.1 *Surface displacement*

When a semiconductor is irradiated by photons with an energy $h\nu$ higher than the band gap (as in our case), electron–hole pairs are generated which thermalize, and eventually the optical excitation is transferred to phonons, allowing to treat the thermal effect as a simple heat source [10]. While the phonons give rise to a thermal expansion, photo-excitation of electron–hole pairs in Si leads to a contraction of the lattice [11]. For irradiation with laser pulses of the order of some 10 ns, it has been found that the second effect dominates and the sample surface opposite to the irradiated side retracts on a typical time scale of a few hundred nanoseconds [11]. We observe the same phenomenon with our set-up, as long as the laser intensity on the Si surface is below the threshold for the formation of a plasma (Fig. 2, trace a). Since the mechanical forces due to acceleration/deceleration of the surface are small in this case because of the rather slow variation of the surface position, this regime is not so relevant for acoustic laser cleaning. When the threshold for the formation of a plasma ($1.1\ \mathrm{J/cm^2}$) is exceeded, however, material expulsion takes place, and the recoil force of the evaporated material leads to a sharp expansion peak of the surface (Fig. 2, traces b and c), with a width comparable to the duration of the laser pulse. The pressure by the expanding plasma is the source of a broader pressure wave through the bulk [12]. For higher intensities this pressure wave outweighs the contraction pulse in Fig. 2(a) and (b) and leads to the "hump" in Fig. 2(c).

In contrast to samples with (100) orientation, for which several pulses corresponding to different acoustic modes are observed, the (111) orientation used here displays only one sharp pulse due to the longitudinal acoustic mode travelling through the sample with a speed of $9.7 \pm 0.1 \times 10^3\ \mathrm{m/s}$, which corresponds very well to the measured sound velocities in (111) silicon [13]. The additional pulses showing up in Fig. 2, traces b and c, at 950 ns are echoes of the original pulses arriving at 315 ns. In Fig. 3 the measured surface displacement at a laser fluence of $1.7\ \mathrm{J/cm^2}$ is plotted on an expanded time scale, together with the acceleration derived from it. The dotted line through the displacement data represents a Savitzky–Golay [14] fit, which was used to obtain a smoothened curve suitable for differentiation. The surface acceleration, calculated from this curve by differentiating twice, displays a sharp maximum–minimum structure with a width of about 5 ns. The maximum acceleration is $0.4 \times 10^9\ \mathrm{m/s^2}$ in this case, the minimum (corresponding to a maximum in deceleration) is $-0.55 \times 10^9\ \mathrm{m/s^2}$. These maxima in acceleration, deceleration and also in the displacement are shown in Fig. 4 as a function of the incident laser fluence F.

The maximum surface acceleration and deceleration increase continuously in the whole investigated fluence range up to $4.5\ \mathrm{J/cm^2}$ and accelerations and decelerations exceeding $10^9\ \mathrm{m/s^2}$ can

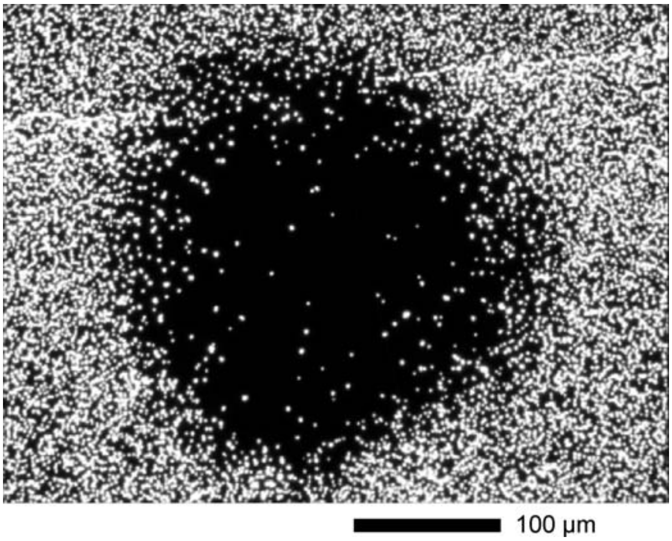

FIGURE 5 Cleaning spot resulting from a bulk acoustic wave, generated by a laser pulse with a fluence of 3 J/cm^2

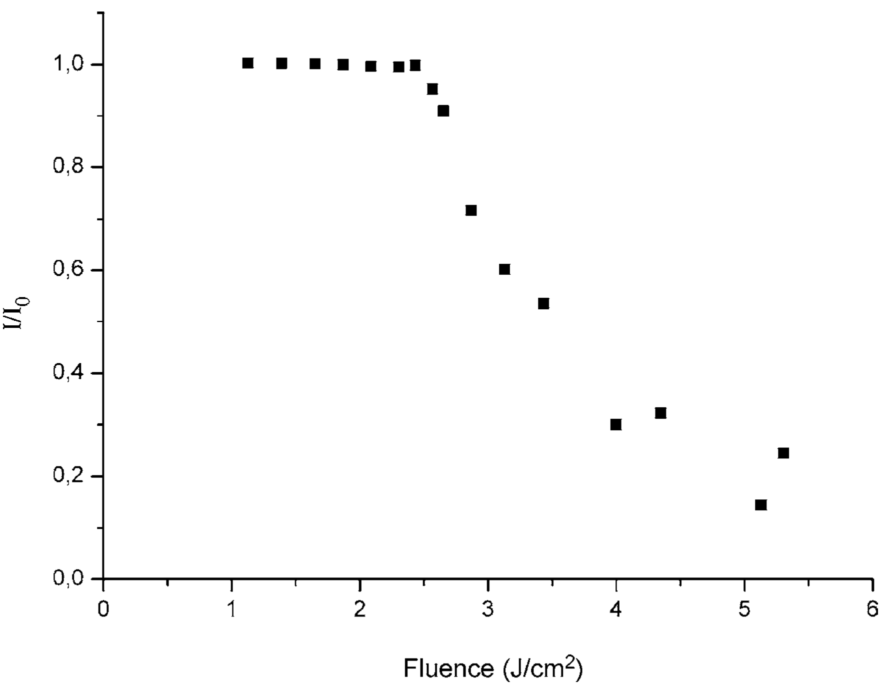

FIGURE 6 Normalized detector signal I/I_0 vs. laser fluence. The threshold value for particle ablation is 2.5 J/cm^2

be reached by the acoustic wave. These relations plotted in Fig. 4 coincide very well with the general dependence of the ablation rate for pulsed laser ablation of solids [15] and the kinetic energy of the expelled ions [16].

More systematic measurements of the surface displacement and the corresponding acceleration as a function of sample thickness, size of the laser spot etc. will be published in a separate paper. However, already the results presented here demonstrate that accelerations (of both signs) in the order of 10^8 g can be reached with this technique, which according to [5] should be sufficient for particles to overcome the adhesion forces and to detach from the sample surface. This will be the topic of the next section.

3.2 *Particle detachment*

In order to study the removal of particles by means of laser-induced acoustic pulses, the Si surface was covered with spherical colloidal test particles. A variety of diameters and materials have been investigated. As an example, we describe here results for 840 nm polystyrene spheres.

Figure 5 represents a dark field image of a surface exposed to an acoustic pulse generated at the back side of the sample by a 3 J/cm^2 laser pulse. The dark spot in the center clearly shows that under these conditions particles are detached indeed. Obviously a few particles inside the spot still stick to the surface, an indication for some scatter in the adhesion forces, which is in line with AFM measurements [5].

The reproducibility of such measurements is demonstrated in Fig. 6. Here the light intensity I scattered by our test particles after the acoustic pulse, normalized by the initial intensity I_0, is plotted versus the fluence of the primary laser pulse (Fig. 6). The reduction of $I/I_0 \sim 1 - \Delta n/n$ at laser fluences above the threshold value $F_c = 2.5\,\text{J/cm}^2$ is a measure of the number of the ablated particles Δn, normalized by n, the total number of particles in the area irradiated by the probe laser. The increase in $\Delta n/n$ with growing laser fluence above the threshold reflects the growing diameter of the ablated spot. This fluence dependence is caused by the small size of the laser-induced plasma in comparison with the sample thickness, and by the related spreading of the acoustic beam as it crosses the sample, so that for increasing fluence the size of the spot with sufficient acoustic intensity to remove the particles gets larger. Comparing the fluence value of the well-defined onset of particle detachment with Fig. 4 reveals that the maximum deceleration at threshold is 0.87×10^8 g. This compares well with estimates on the basis of the simple picture that detachment sets in when the inertial force $F_{\text{in}} = ma$ of the particle exceeds the adhesion force F_{adh}. We can determine the adhesion forces of 840 nm with this simple picture to 282 nN, which appears to agree reasonably well well with the AFM data [5] of 150 nN.

However, a closer inspection shows that this simple picture has to be modified, because an additional mechanism known as the "trampoline effect" [3] comes into play. Evidence for this phenomenon comes from a measurement of the velocity v_p of the detached particles, using an optical time-of-flight technique [17]. The resulting values for

v_p were found to be in the order of 20 m/s, whereas the maximum velocity of the surface as taken from Fig. 4 at a fluence of 2.5 J/cm^2 is only 1.7 m/s. This at first sight paradoxical result can be explained by taking into account that during the initial acceleration phase of the surface the particle is elastically deformed, and that the stored elastic energy is partially converted into kinetic energy during the detachment process. Thus for a quantitative analysis of our results a more detailed nanomechanical model has to be developed. Such work is under way.

4 Conclusions

In conclusion, we have shown that acoustic laser cleaning, based on a laser-induced acoustic wave generated at the back side of the sample, can be used to detach small particles from surfaces. Due to the separation of optical and acoustical effects, this technique is much better suited for studying the basics of the detachment process than conventional dry laser cleaning, where it is difficult to distinguish between mechanical and optical near field effects. A fast, sensitive interferometer has been developed which allows us to determine the displacement (and therefore also the velocity and acceleration) of the sample surface with high accuracy, so that the essential input parameters are known to develop a model for the detachment process. In addition to possible cleaning applications this method should also allow – using a proper model for the detachment – determination of adhesion forces of small particles on short time scales, which experimentally have not been accessible so far, and which are complimentary to adhesion force measurements with the atomic force microscope.

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